Fluctuations in the aging dynamics of structural glasses

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Motivation

- Experiments show spatially heterogeneous dynamics.
- Goal: understanding spatial fluctuations in glassy dynamics.

Outline

1. Experiments show strong fluctuations (“dynamic heterogeneities”) in the aging regime.

2. Soft mode approach to local fluctuations: the age of the sample fluctuates locally. Scaling predictions and simulation results for local fluctuations in a spin glass.

3. Scaling in the behavior of local fluctuations in a structural glass: probability distributions of one-point, two-time observables.

4. Scaling in the behavior of local fluctuations in a structural glass: spatial correlations.

5. Scaling in the behavior of local fluctuations in a structural glass: crossover between aging and equilibrium regimes.

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The Problem: Dynamical heterogeneities

Colloid: confocal microscopy (Courtland and Weeks, J. Phys. Cond. Mat **15** S359 (2003))

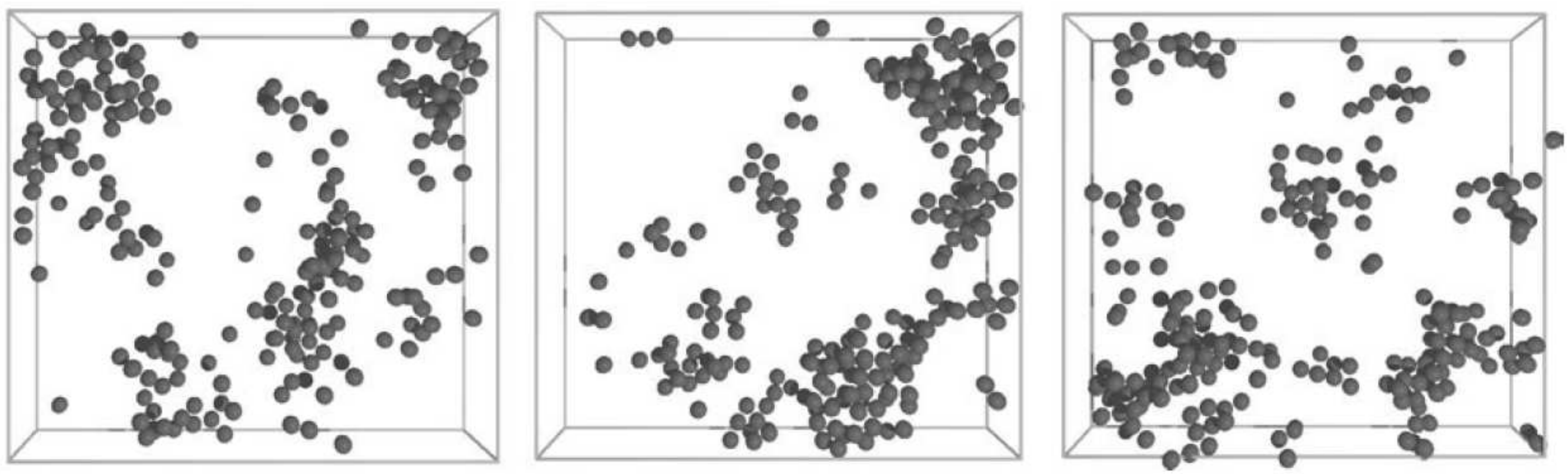
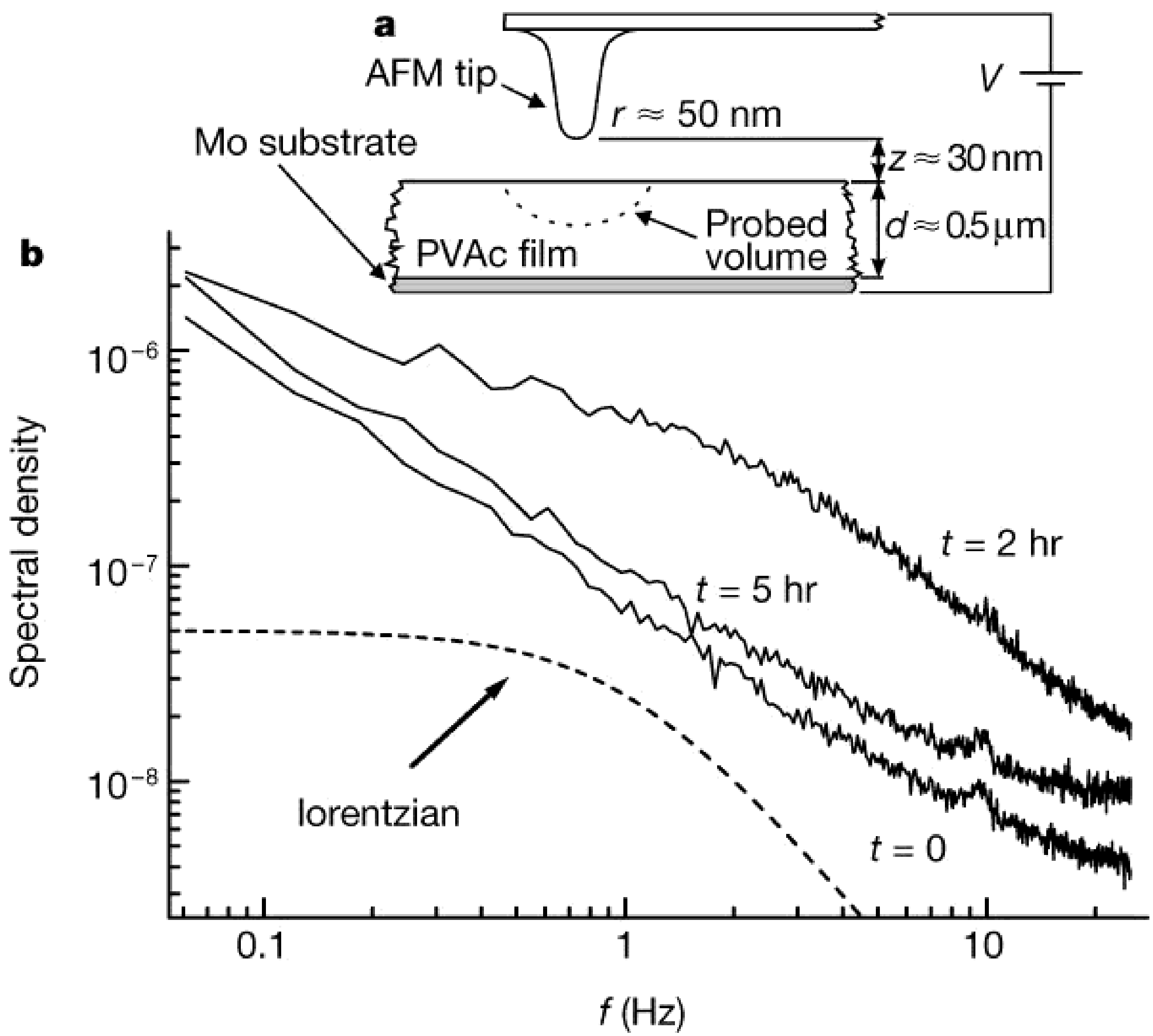


Figure 4. Locations of the 10% most mobile particles at three different ages t_w. For each picture, mobility was determined by calculating displacements Δr over an interval $[t_w, t_w + \Delta T]$, with $\Delta T = 10$ min. Left: $t_w = 10$ min, and $\Delta r > 0.43$ μm for the most mobile particles. Middle: $t_w = 55$ min, $\Delta r > 0.34$ μm. Right: $t_w = 95$ min, $\Delta r > 0.33$ μm. The data are the same as shown in previous figures, and the choices of t_w correspond to local maxima of γ in figure 2(a). The particles are drawn to scale (2.36 μm diameter) and the box shown is the entire viewing volume (within a much larger sample chamber).

The Problem: Dynamical heterogeneities

PVAc: dielectric fluctuations (Vidal Russell & Israeloff, Nature **408**, 695 (2000))



Polymer glass, $T = T_g - 9K$, transient appearence of strongly fluctuating region under tip

Heterogeneity lifetime ≈ relaxation time

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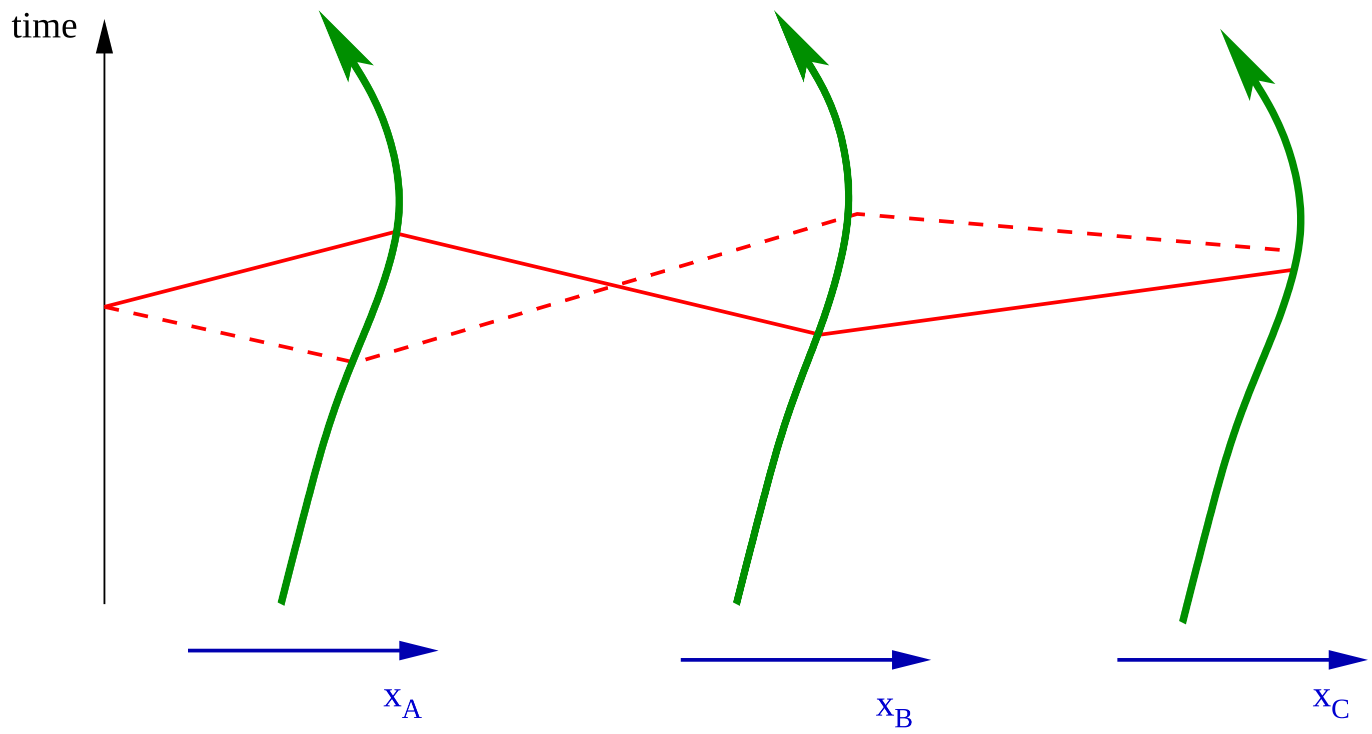
Can we understand dynamical heterogeneities in aging systems?

A possible explanation: the glassy material is aging, but the ages are fluctuating in space.



RG in time: reparametrizations $t \to h(t)$ leave "dynamical action" $\mathcal{S}$ unchanged (irrelevant terms break symmetry at finite times) (C.Chamon, M.P.Kennett, H.E.C., L.F.Cugliandolo, PRL **89**, 217201 (2002))

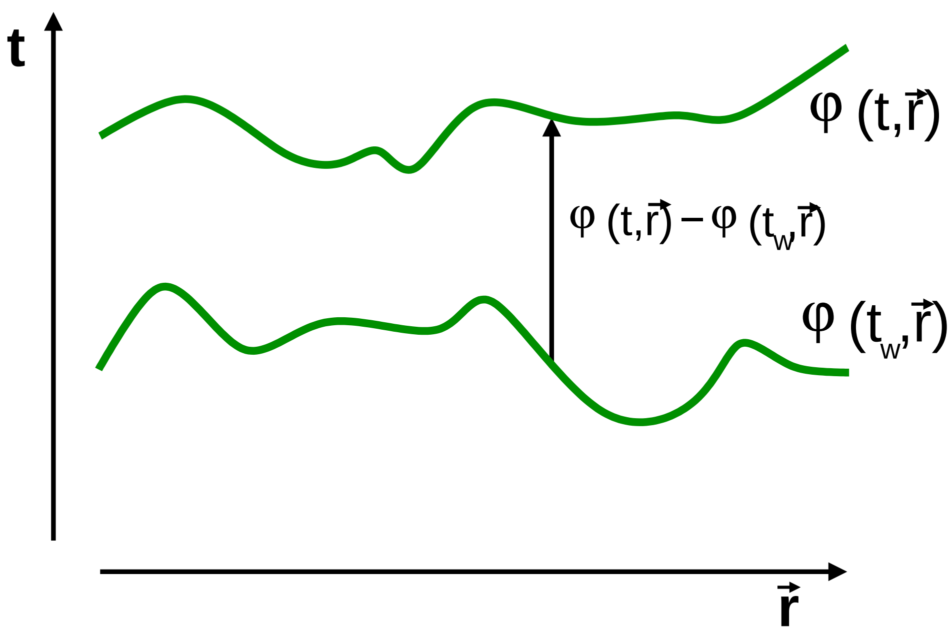
Probability distribution of local correlations: $\rho(C_{\vec{r}})$

(with C. Chamon, L. Cugliandolo, J. Iguain, and M. Kennett: PRL **88**, 237201 (2002) and PRB **68**, 134442 (2003))

If $C_0(t,t_w) \approx C_0(h(t)/h(t_w))$ (for example, $h(t) \approx t$ in 3DEA) then:

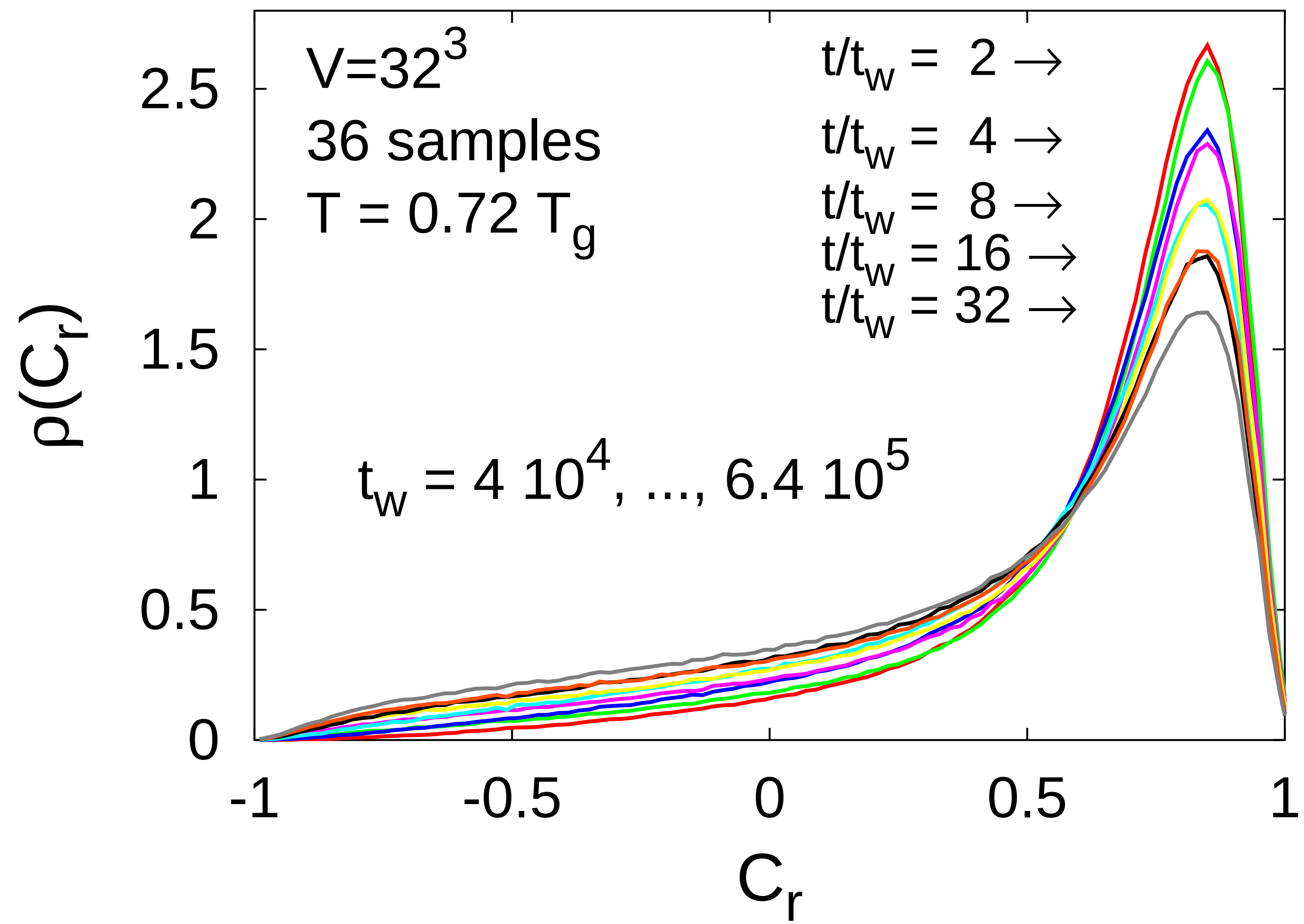
$$t \to h_{\vec{r}}(t) = \mathrm{e}^{\varphi_{\vec{r}}(t)}$$

$$C_{\vec{r}}(t,t_w) = C_0(h_{\vec{r}}(t)/h_{\vec{r}}(t_w)) = C_0(\exp(\varphi_{\vec{r}}(t) - \varphi_{\vec{r}}(t_w)))$$



- Fluctuating $\varphi_{\vec{r}}(t)$
- Time reparametrization invariance
- $\Rightarrow \varphi_{\vec{r}}(t) - \varphi_{\vec{r}}(t_w) \approx$ $\ln\left(\frac{h(t)}{h(t_w)}\right) + \sqrt{a + b\ln\left(\frac{h(t)}{h(t_w)}\right)}\, X_r$

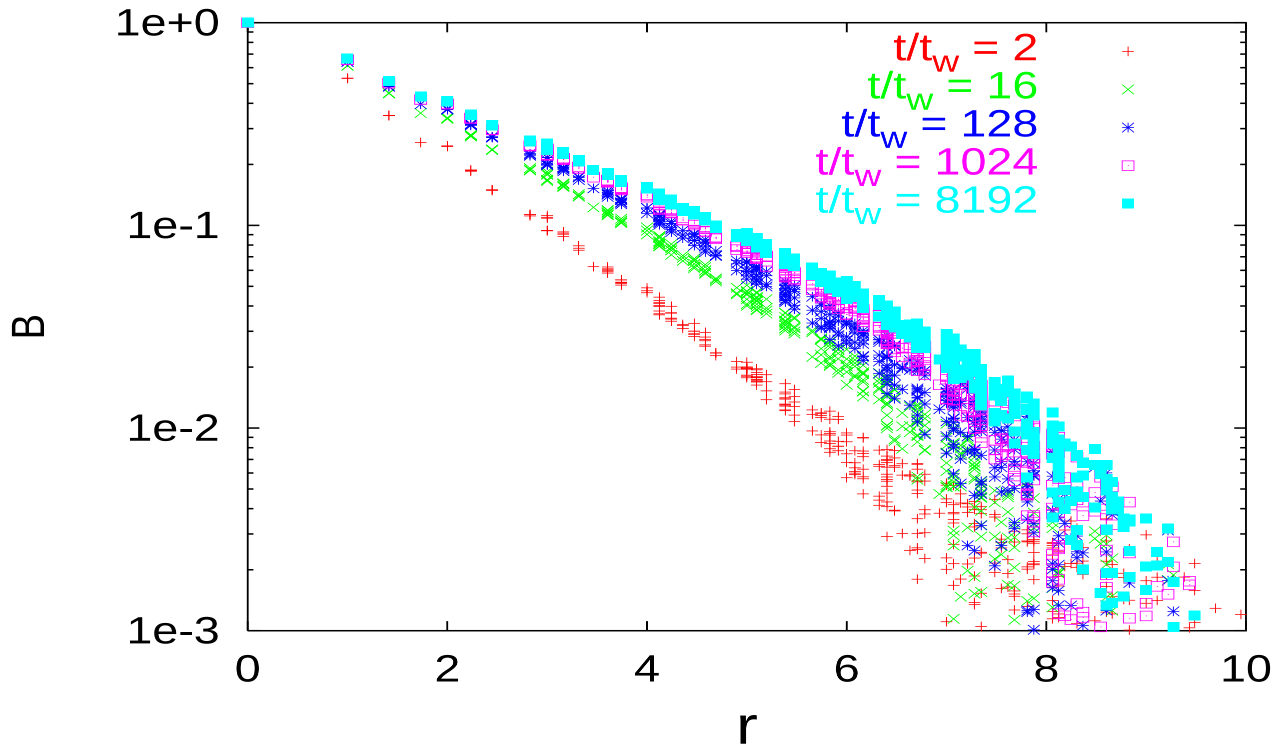
Collapse of $\rho(C_{\vec{r}})$ for fixed t/t_w



Noise-noise spatial correlations: exponential decay

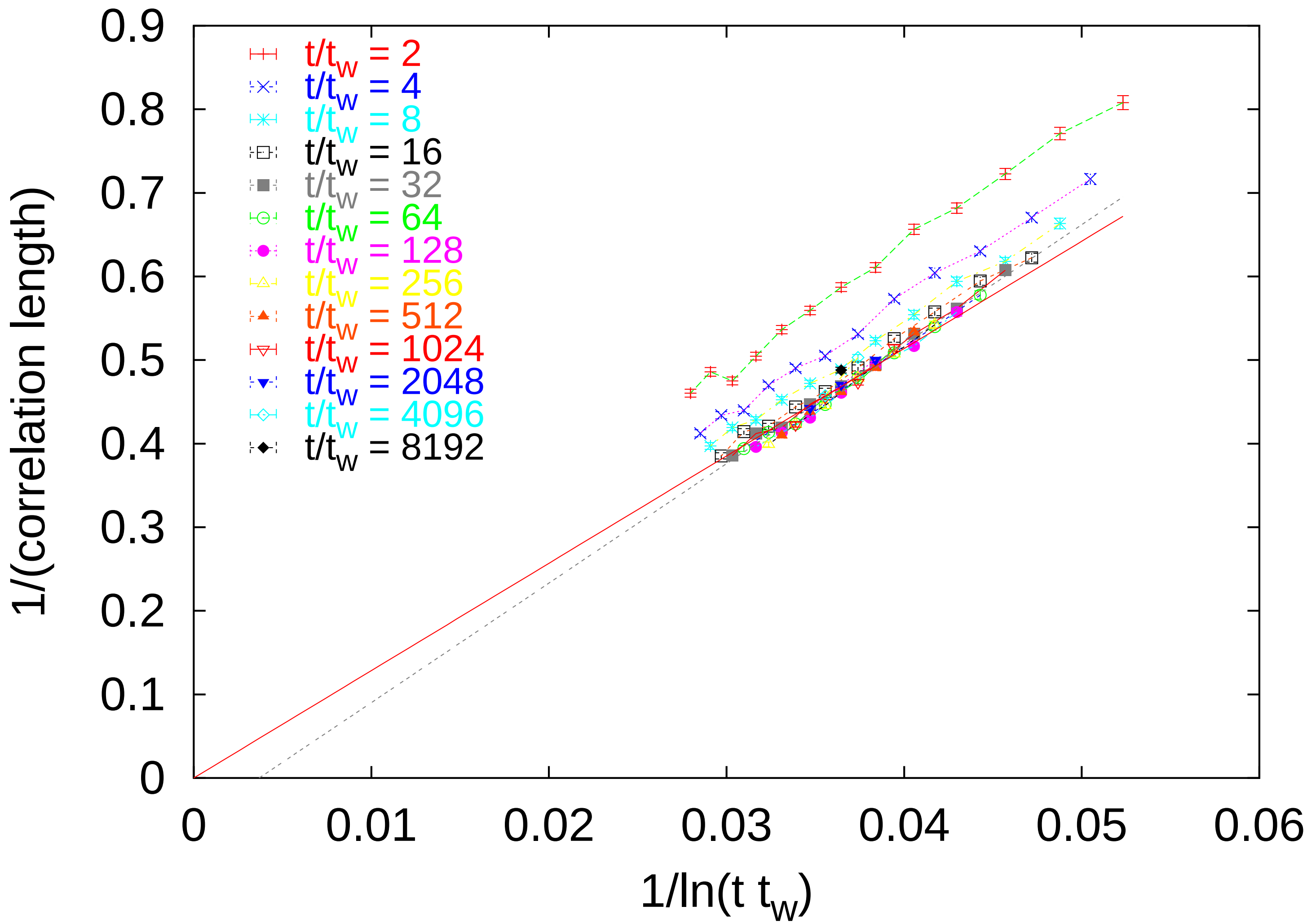
RG-irrelevant symmetry-breaking terms $\Rightarrow$ expect finite correlation length ($\to \infty$ for infinite t, t_w).

$$B(\vec{r}, t, t_w) \equiv \langle \delta C_{\vec{r}_i}(t, t_w)\ \delta C_{\vec{r}_i + \vec{r}}(t, t_w) \rangle_{\vec{r}_i}$$



$t_w = 10^4$ MCs, $V = 32^3$, $T = 0.72 T_g$, 64 disorder realizations

Correlation length $\xi(t, t_w) \rightarrow \xi(t t_w)$



$V = 32^3$, $T = 0.72 T_g$, 64 disorder realizations

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3. **Scaling in the behavior of local fluctuations in a structural glass: probability distributions of one-point, two-time observables.** (HC and A. Parsaeian, Nature Physics 2007)

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5. Scaling in the behavior of local fluctuations in a structural glass: crossover between aging and equilibrium regimes.

Structural glass simulations

- 80:20 binary Lennard-Jones mixture, 8000 particles. Thermalized at $T_i = 5.0$, time origin at instantaneous quench to $T_f = 0.4$ (below $T_g \approx 0.435$). Evolves for up to 100000 LJ units (i.e. $\sim 10^{-8}s$) after quench. β relaxation time is of the order of 1 LJ unit. Repeated for 250 to 4000 independent runs (depending on timescale).

- Divide the system in regions, and measure one point, two time quantities for each region.

  $$C_{\vec{r}}^{\text{part}}(t, t_w) \equiv \frac{1}{\mathcal{N}(V_{\vec{r}})} \sum_{\vec{r}_i(t_w) \in V_{\vec{r}}} \cos(\vec{q} \cdot [\vec{r}_i(t) - \vec{r}_i(t_w)])$$

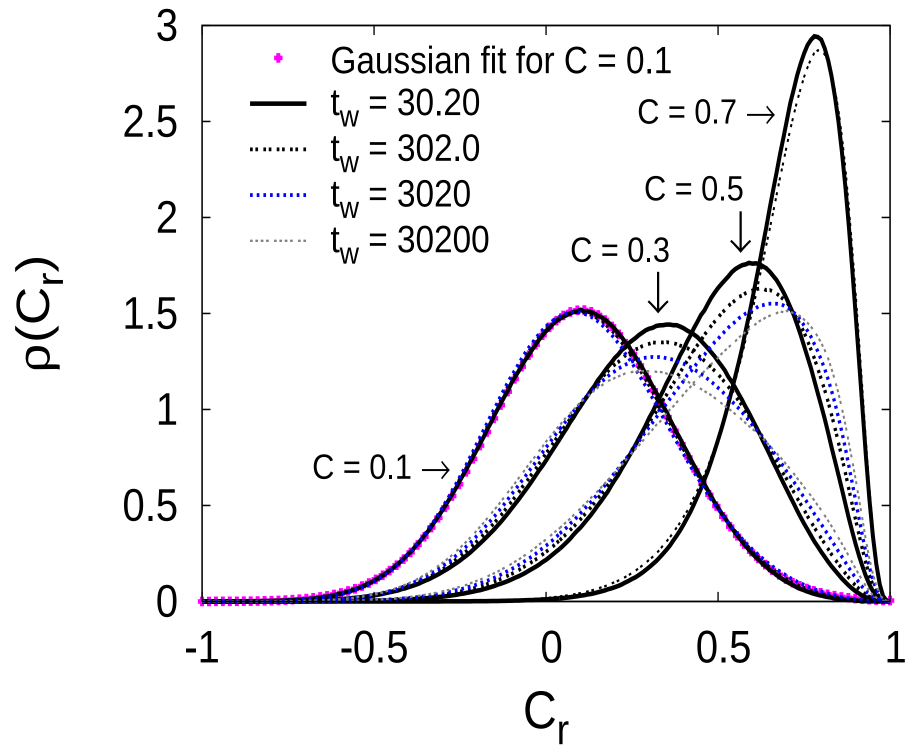
  Obtain the probability distributions $\rho(C_r)$ for the local values.

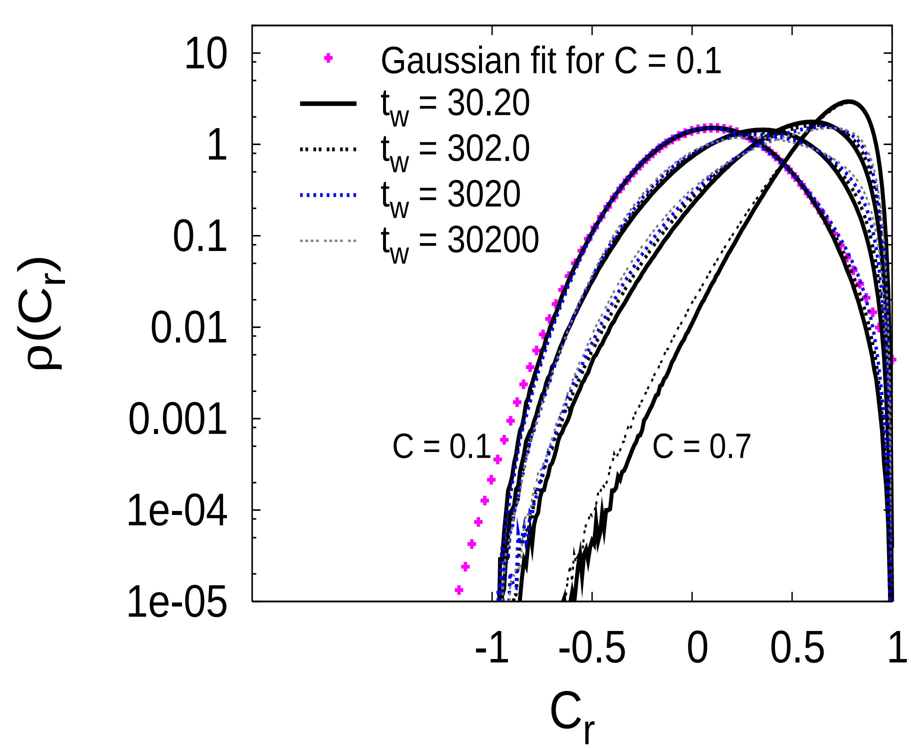
- Use the *global* intermediate scattering function

  $$C_{\text{global}}(t, t_w) \equiv \frac{1}{N} \sum_{i=1}^{N} \cos(\vec{q} \cdot [\vec{r}_i(t) - \vec{r}_i(t_w)])$$

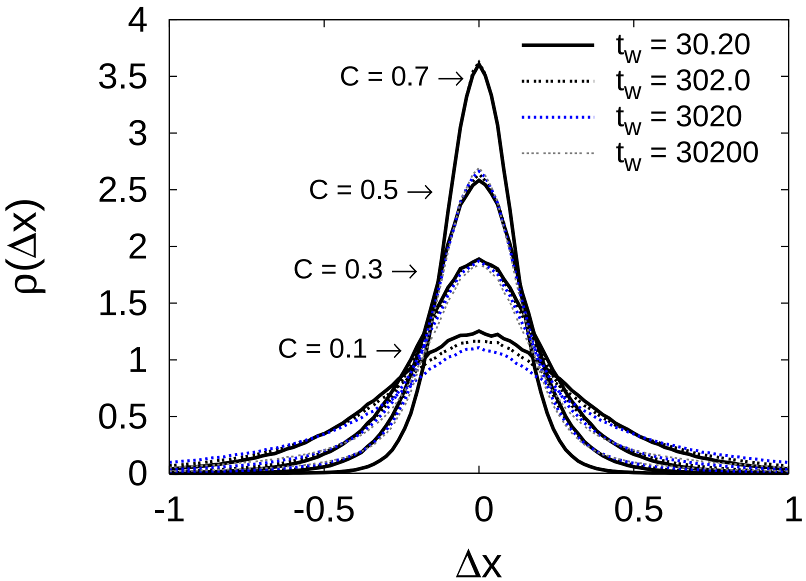
  to quantify how correlated the system is between times t_w and t.

Approximate collapse of $\rho(C_r)$ at constant $C_{\text{global}}(t, t_w)$

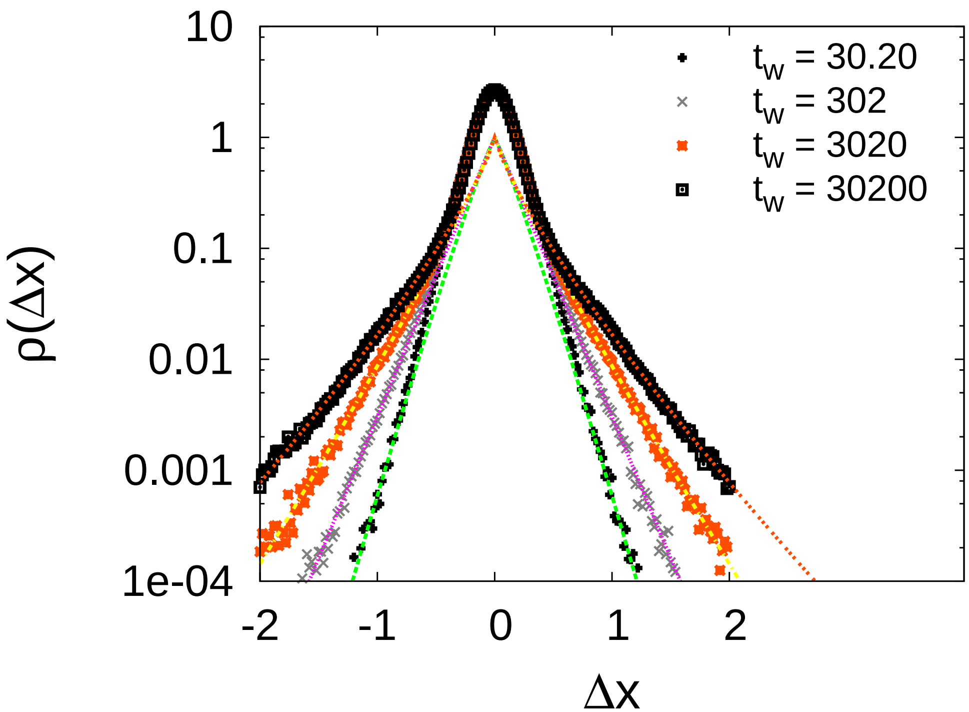




Distribution of one-dimensional displacements $\rho(\Delta x)$

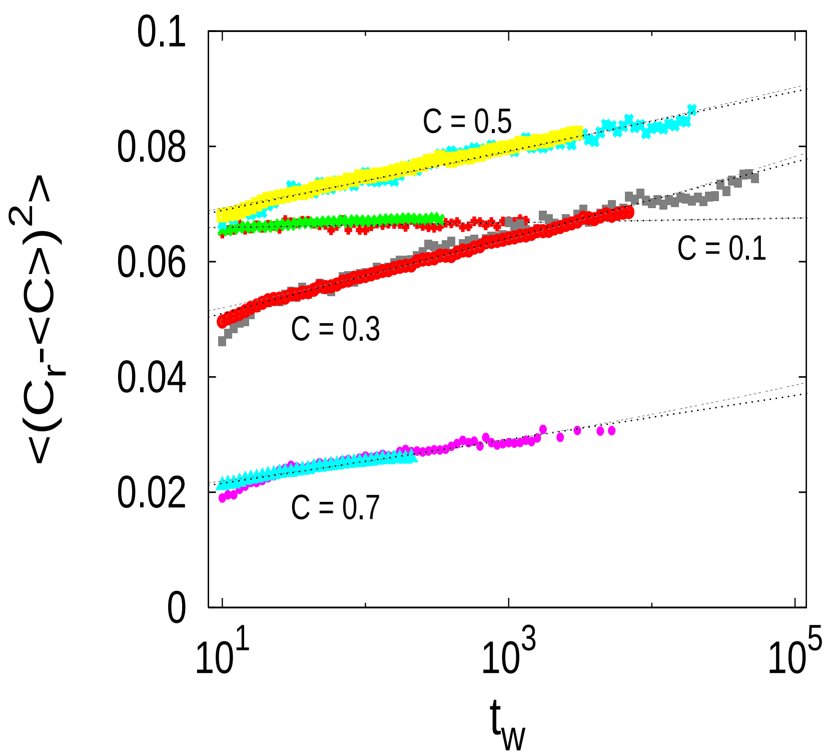


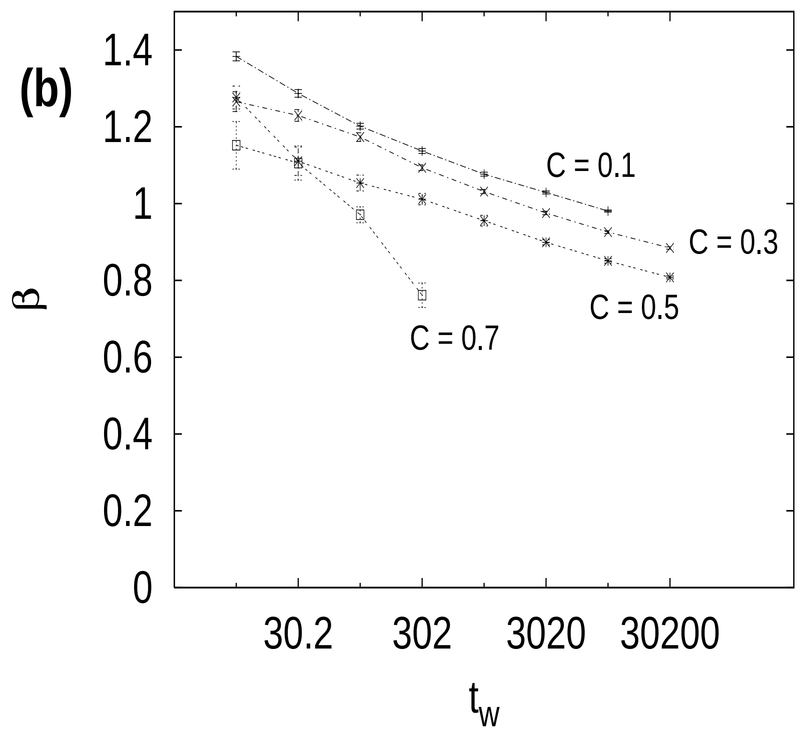
approximate collapse at constant $C_{\text{global}}(t, t_w)$.



nonlinear exponential tails
β exponent decreases for increasing t_w

Slow t_w dependences at constant $C_{\text{global}}(t, t_w)$





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Dynamical correlations: densities

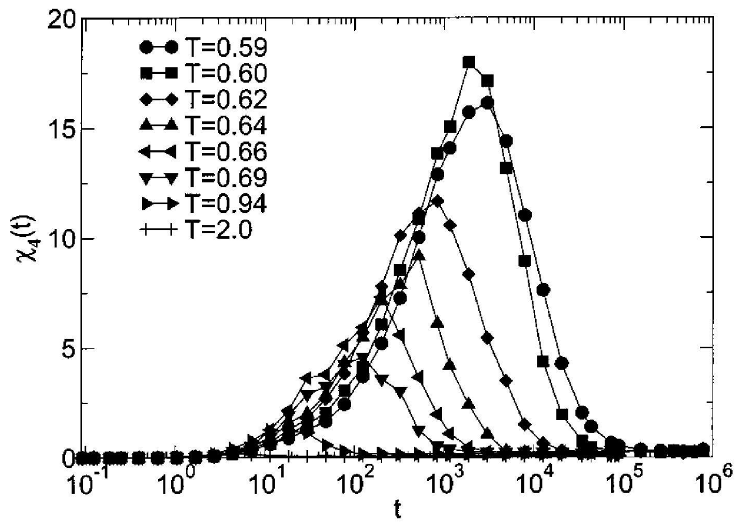
(Lačević, Starr, Schrøder, Glotzer J. Chem. Phys **119**, 7372 (2003))

$$
\begin{aligned}
w(\mathbf{r},t,t_w) &= \text{1 if particle at } \mathbf{r} \text{ has moved} < a_{\text{vib}} \\
&= \text{0 otherwise} \\
g_4(\mathbf{r},t,t_w) &= \text{spatial correlation of } w(\mathbf{r},t,t_w) \\
\xi_4(t,t_w) &= \text{correlation length for } g_4(\mathbf{r},t,t_w) \\
\chi_4(t,t_w) &= \text{dynamic density susceptibility} \\
&\propto \int d^3r \; g_4(\mathbf{r},t,t_w)
\end{aligned}
$$

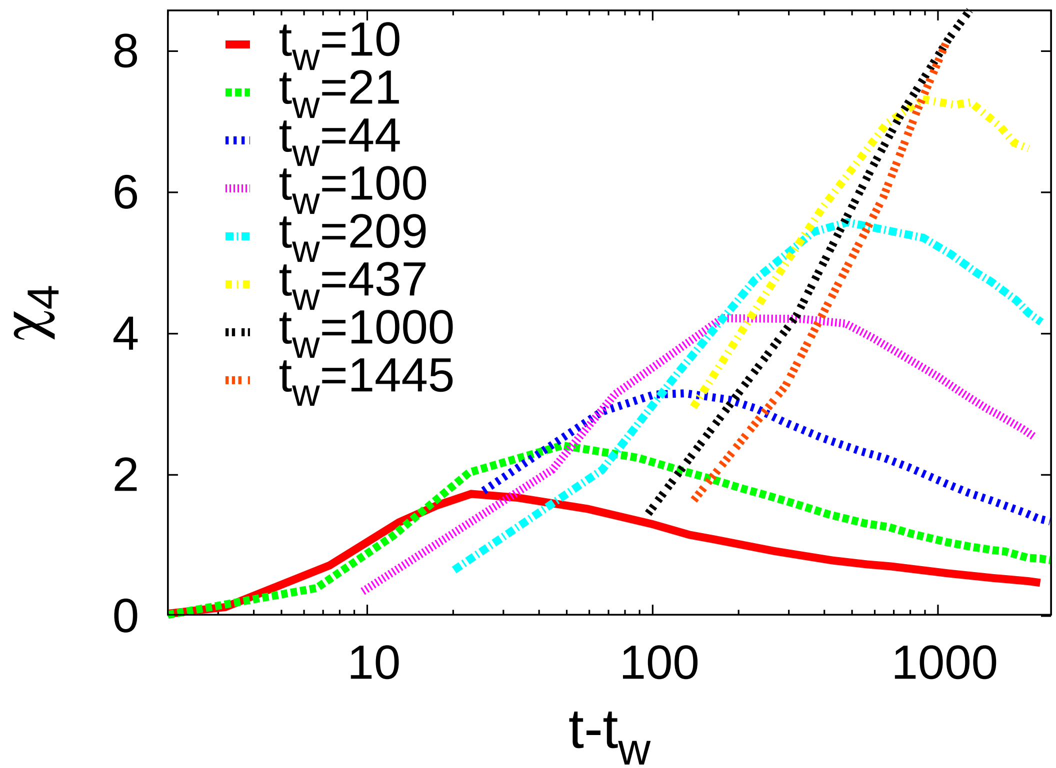
Time evolution of χ_4

Supercooled regime

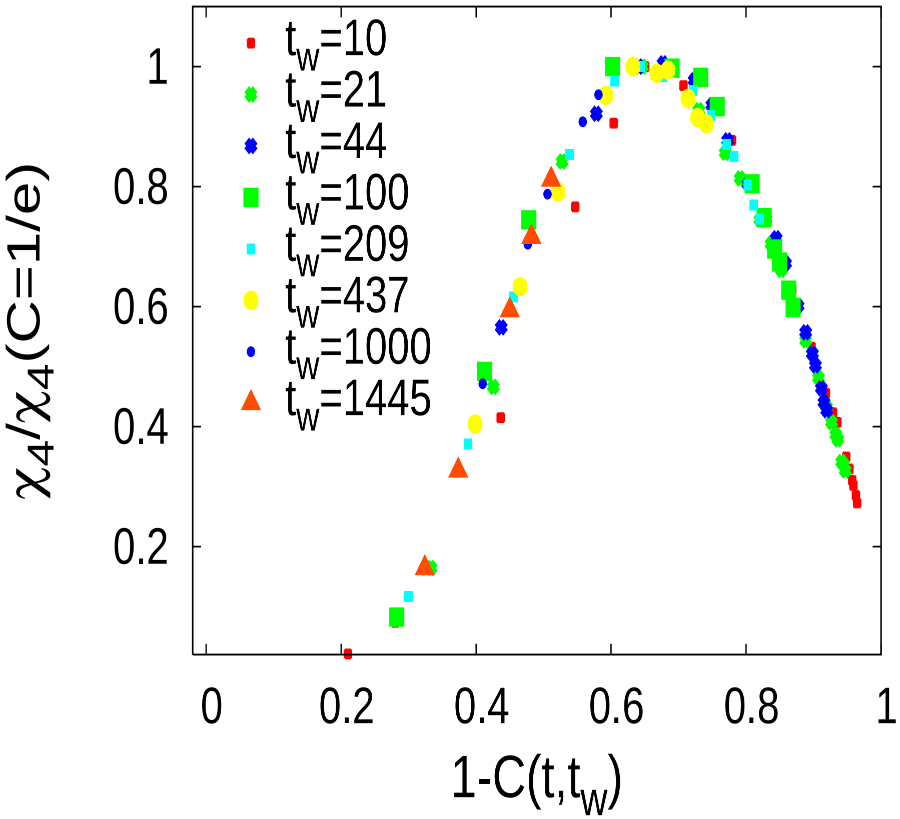
(Starr et al. J. Chem. Phys. 2003)

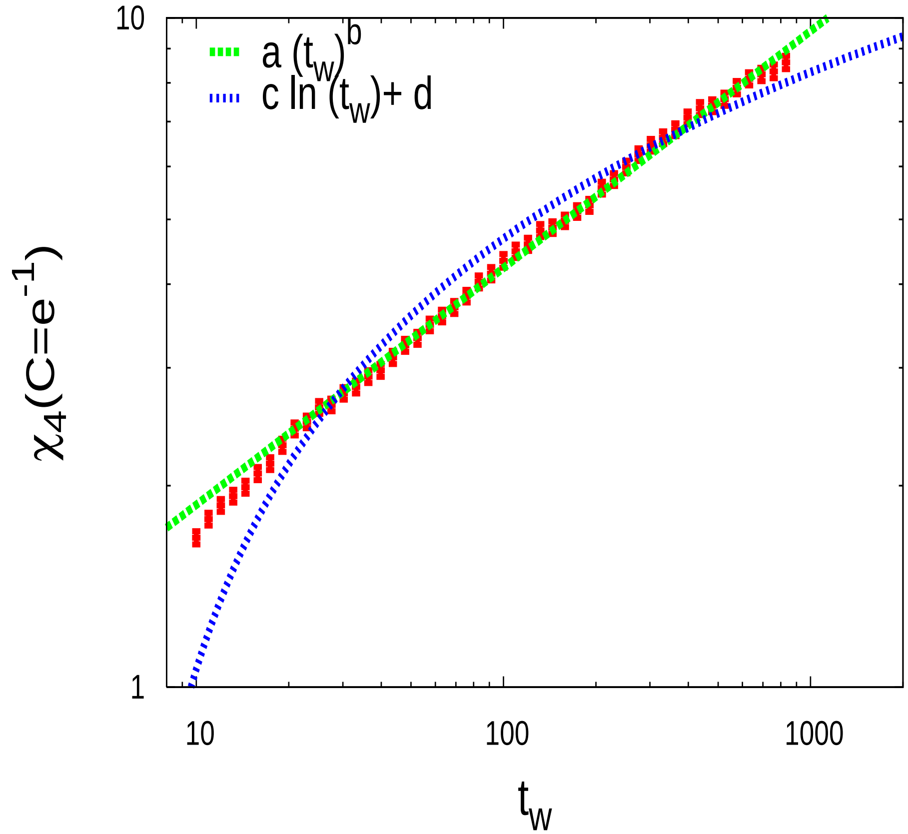


Aging regime



Scaling of χ_4



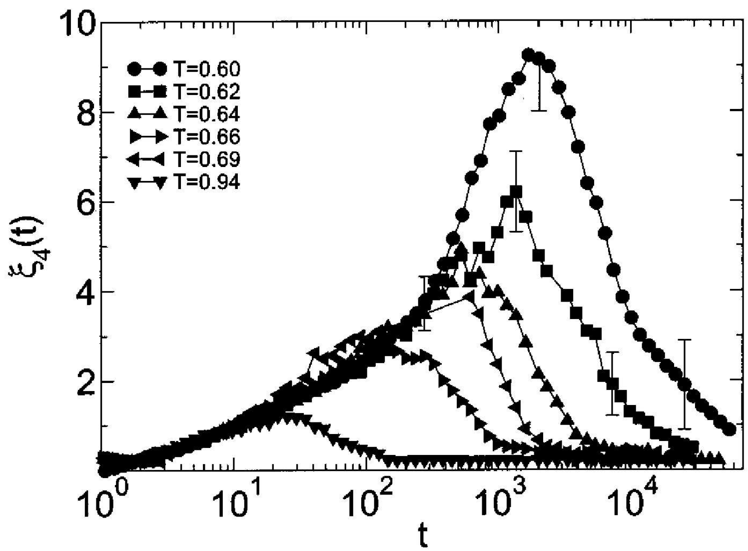


$b = 0.353 \pm 0.003$

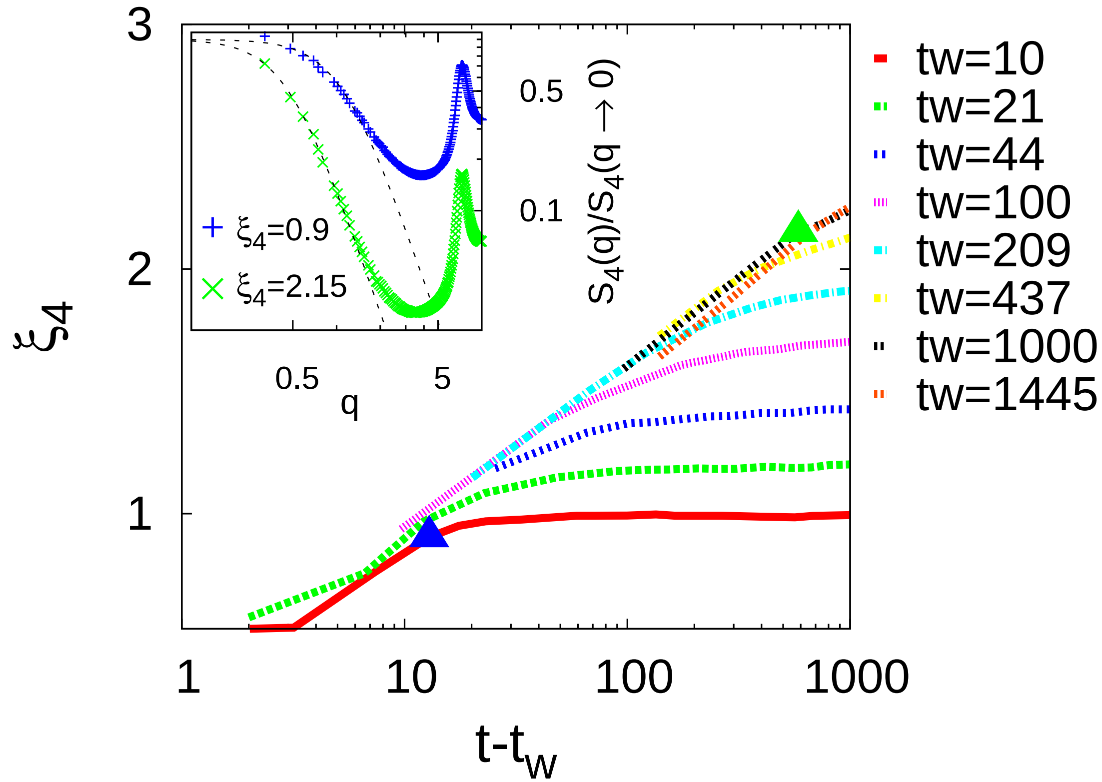
Time evolution of ξ_4

Supercooled regime

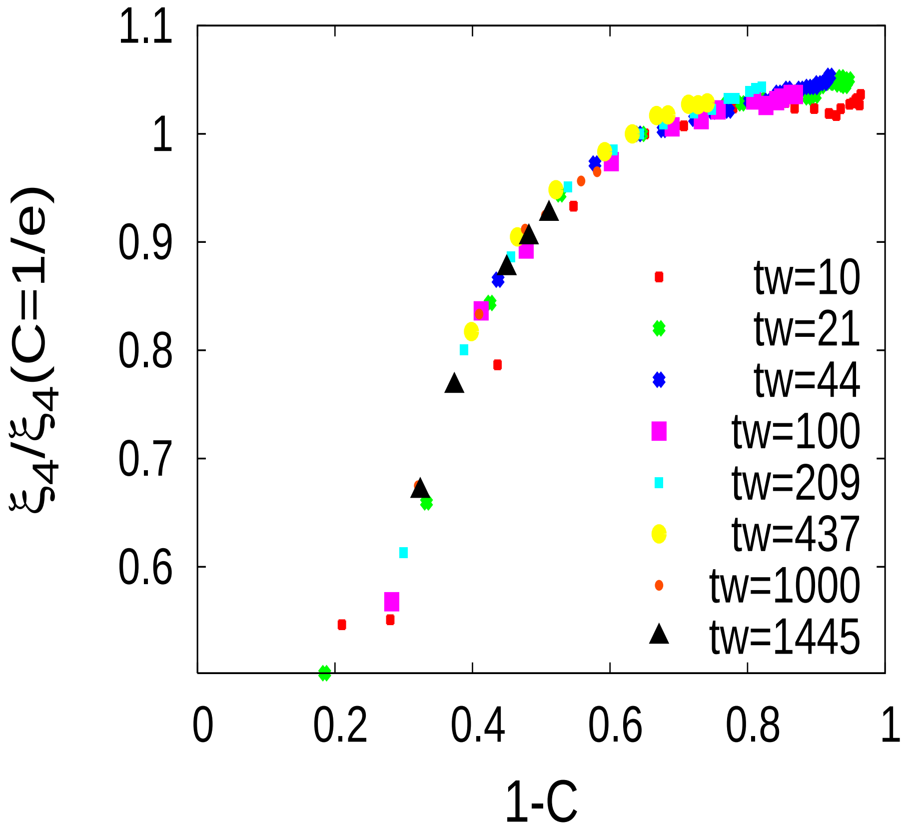
(Starr et al. J. Chem. Phys. 2003)

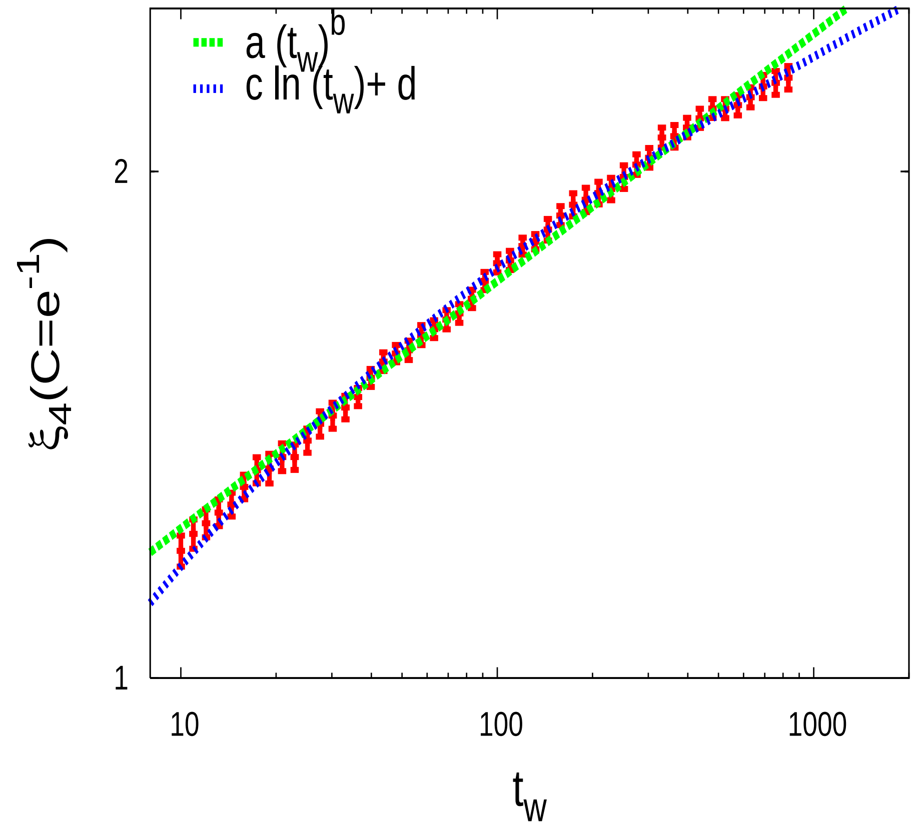


Aging regime



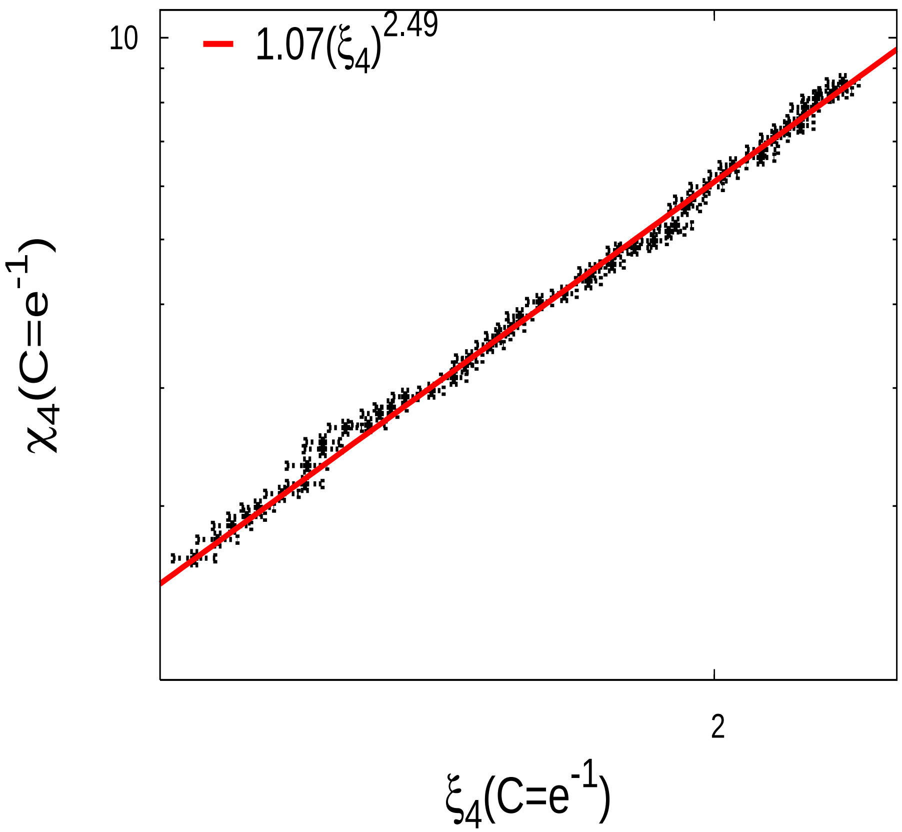
Scaling of ξ_4



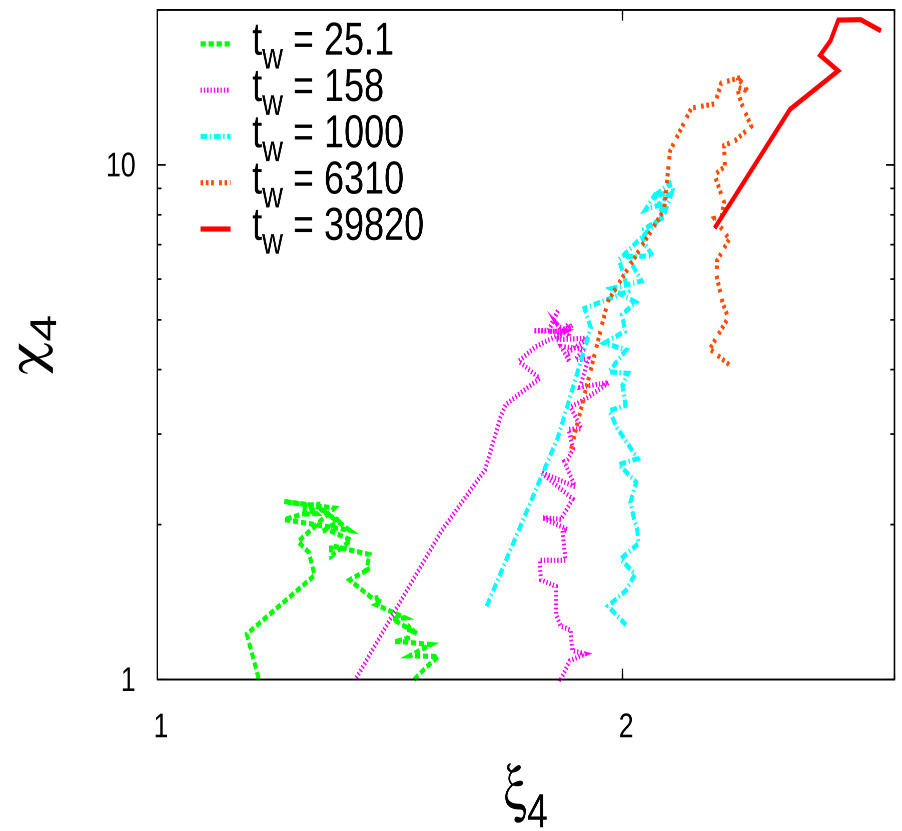


$b = 0.146 \pm 0.001$

χ_4 vs. ξ_4



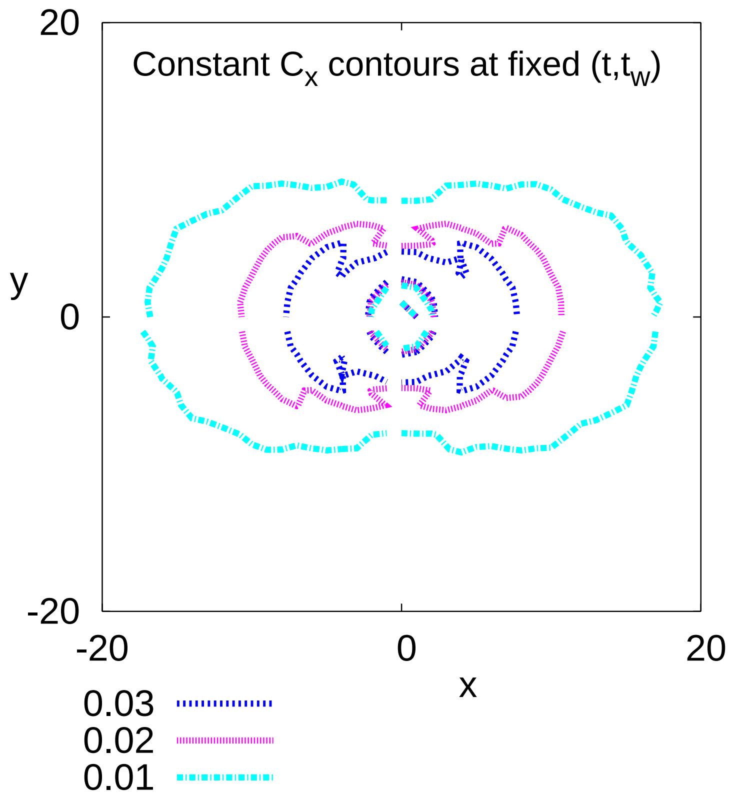
1.07(ξ4)^2.49
10
2
χ4(C=e^-1)
ξ4(C=e^-1)



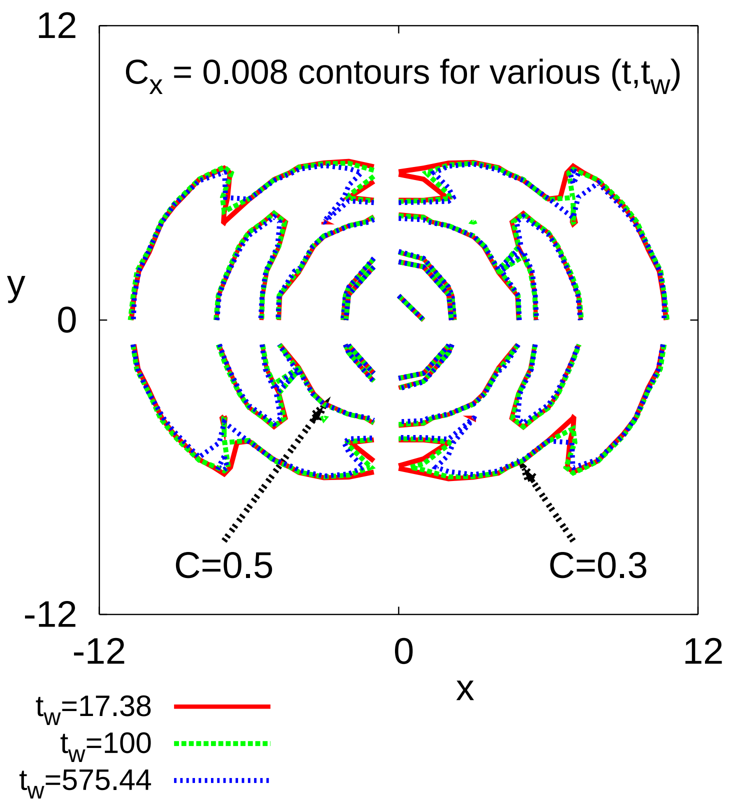
tw = 25.1
tw = 158
tw = 1000
tw = 6310
tw = 39820
10
1
1
2
χ4
ξ4

Anisotropy

$$C_x(\mathbf{R}, t, t_w) \equiv \langle \delta x(\mathbf{R}, t, t_w) \delta x(\mathbf{0}, t, t_w) \rangle_c$$



$tw = 30.2, t = 2511.88$



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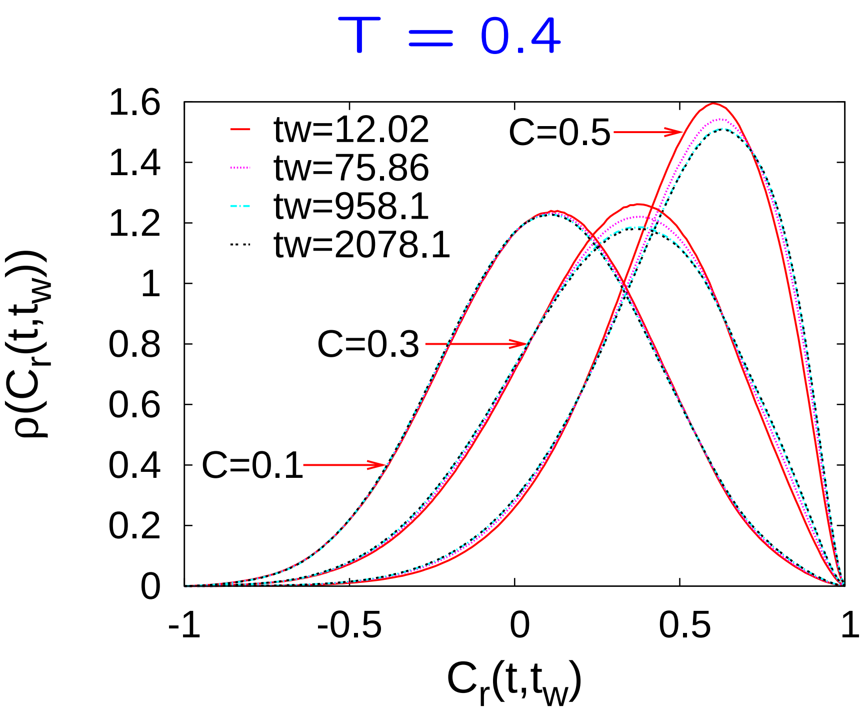
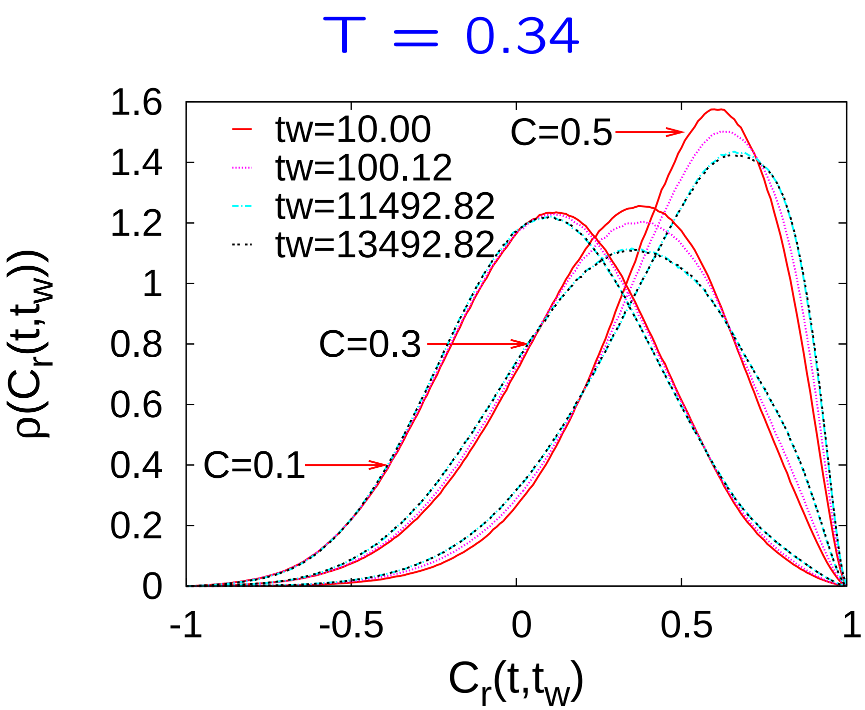
Fluctuations: crossover from aging to equilibrium

WCA potential (repulsive only)

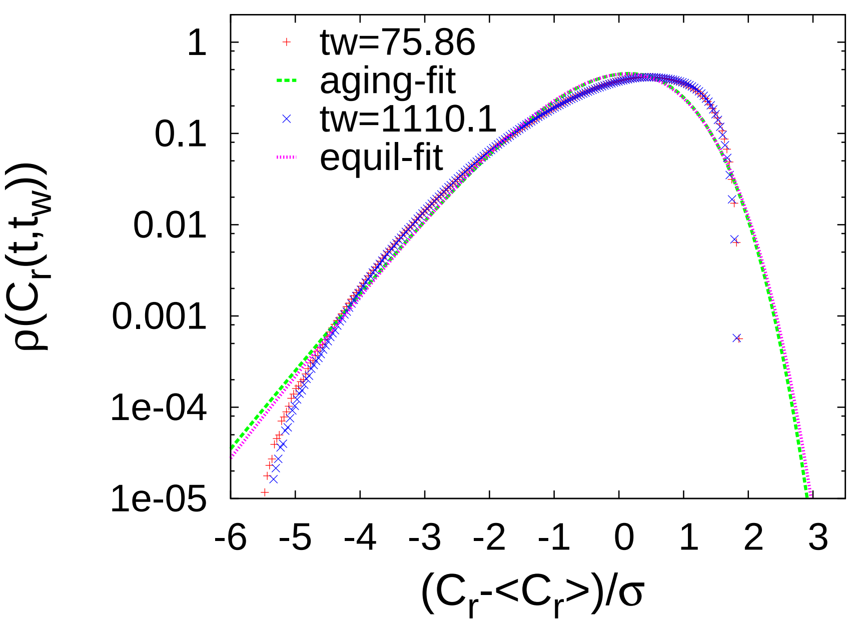




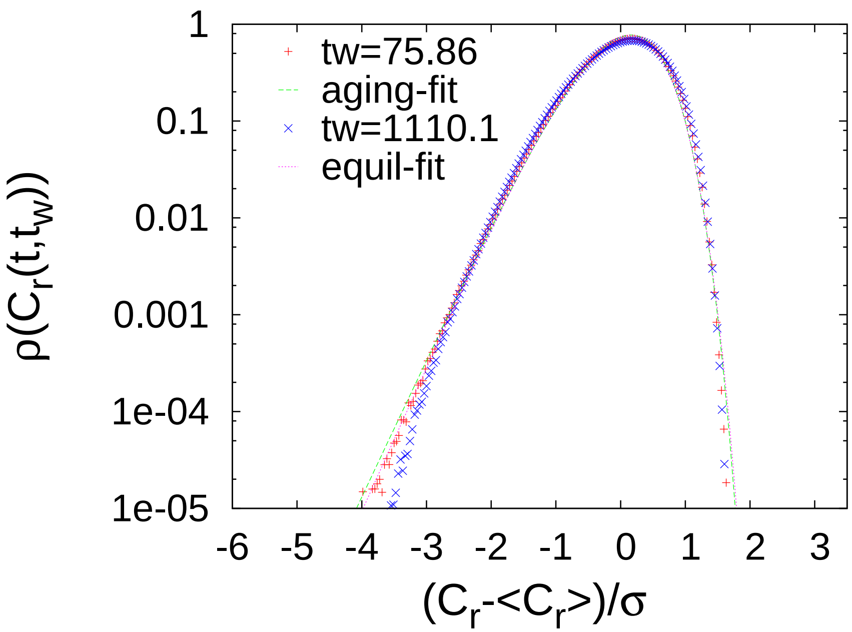
Fluctuations: crossover from aging to equilibrium

WCA potential (repulsive only)

coarse graining: 6.6 particles



coarse graining: 30.5 particles



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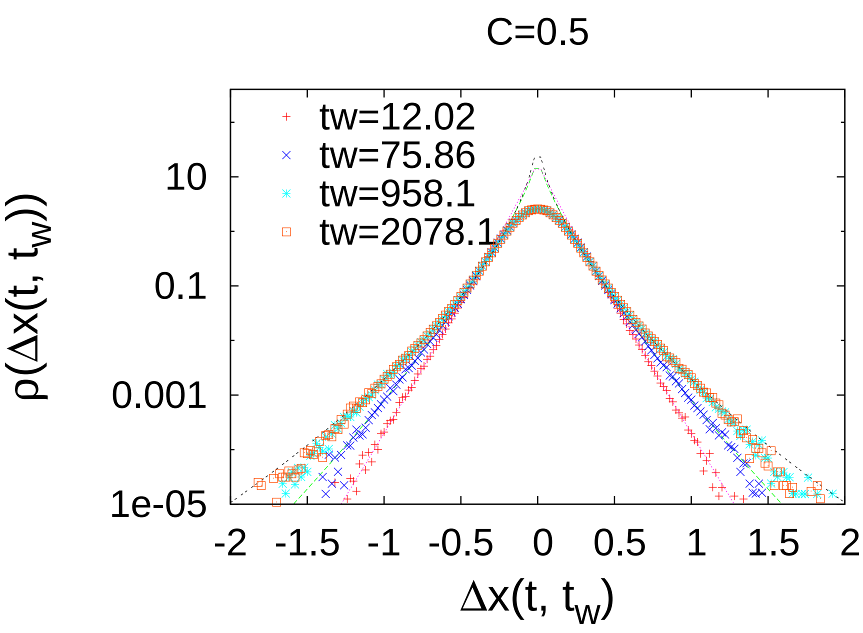

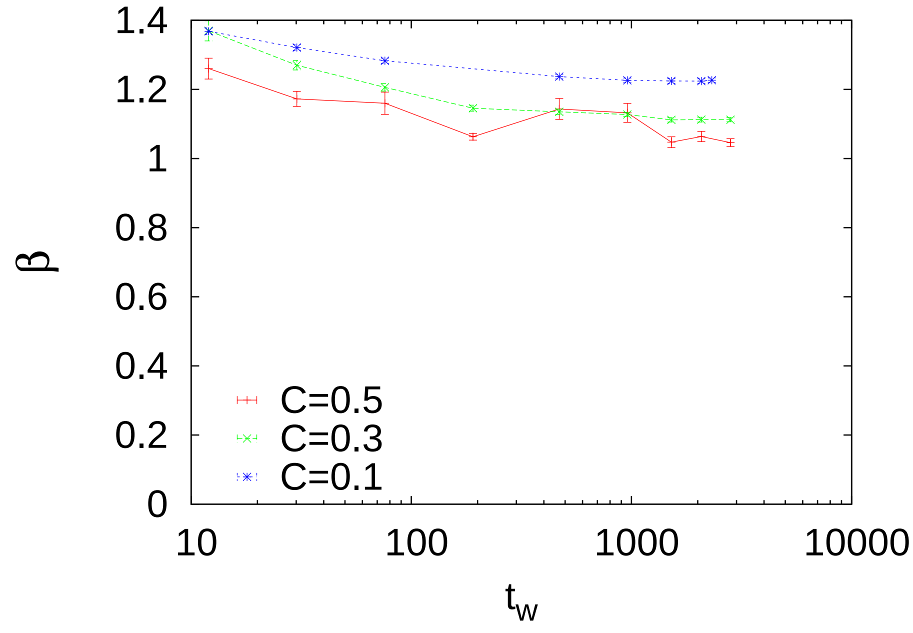

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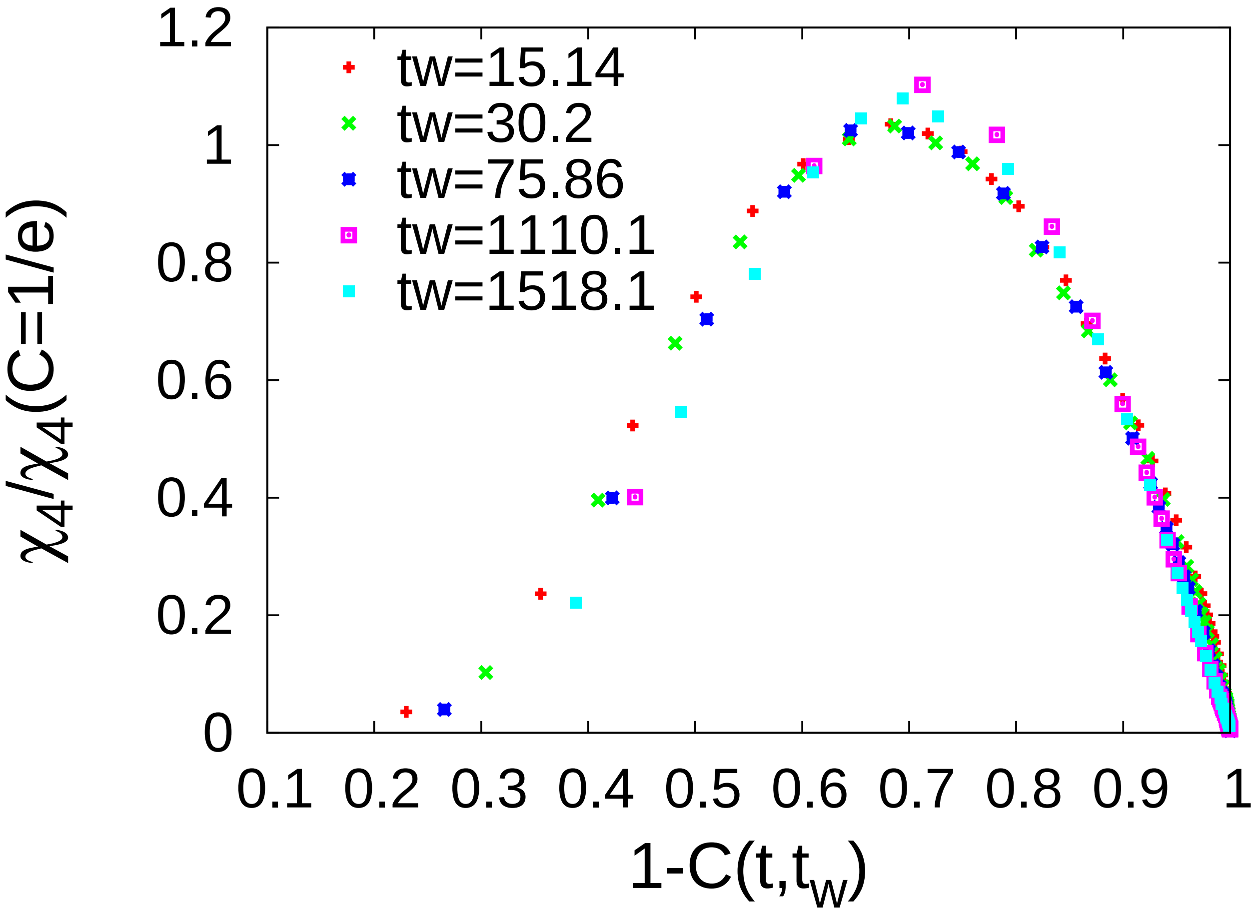

Summary

- Aging in a binary LJ: Probability distributions of local two-time quantities like C_r and Δx show approximate collapse at fixed $C_{\text{global}}(t, t_w)$. Slow drift of distributions with t_w, no timescale observed. Tails of $\rho(\Delta x)$ are nonlinear exponential with exponent $\beta \approx 0.8 - 1.4$, with the lower β corresponding to the longest t_w.

- Aging in a binary LJ: Scaling of 4-point density correlation $\chi_4(t, t_w) \approx {\chi_4}^0(t_w)\phi(C(t, t_w))$, with $\lim_{C(t,t_w)\to 0} \phi(C(t, t_w)) = 0$. Scaling of the correlation length $\xi_4(t, t_w) \approx {\xi_4}^0(t_w)\varphi(C(t, t_w))$, with $\lim_{C(t,t_w)\to 0} \varphi(C(t, t_w)) = \varphi_0 \neq 0$. Data are consistent with a power law $\chi_4(C = 1/e) \sim (\xi_4(C = 1/e))^b$, however the decay of $\chi_4(t, t_w)$ when $C(t, t_w) \to 0$ does not correspond to a decay in $\xi_4(t, t_w)$.

- Aging and equilibrium in a binary WCA: One-point distributions seem identical in the aging and equilibrium regimes. (Collapse is better when small coarse graining regions are used due to correlation length effects). The relationship between the rescaled χ_4 and $C(t, t_w)$ is also the same in the aging and equilibrium regimes.

Determination of ξ_4

Fit $S_4(q,t,t_w)$ at small q using the form: $S_4(q,t,t_w) = \frac{a}{1+(\xi_4 q)^\gamma} + b$.